

FORM PTO-1390 (REV. 12-2001)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER 5294-000010 U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <b>10/088186</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			
INTERNATIONAL APPLICATION NO. PCT/KR00/01039	INTERNATIONAL FILING DATE 15 September 2000 (15.09.00)	PRIORITY DATE CLAIMED 15 September 1999 (15.09.99)	
TITLE OF INVENTION A NOVEL AROMATIC POLYSULFIDE AND AN ASPHALT COMPOSITION CONTAINING THE SAME			
APPLICANT(S) FOR DO/EO/US SHIM, Kyung-Sup			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.</p> <p>4. <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p>a. <input type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau).</p> <p>b. <input checked="" type="checkbox"/> has been communicated by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p>a. <input checked="" type="checkbox"/> is attached hereto.</p> <p>b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).</p> <p>b. <input type="checkbox"/> have been communicated by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p><b>Items 11 to 20 below concern document(s) or information included:</b></p> <p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input type="checkbox"/> A <b>FIRST</b> preliminary amendment.</p> <p>14. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</p> <p>15. <input type="checkbox"/> A substitute specification.</p> <p>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</p> <p>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</p> <p>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</p> <p>20. <input checked="" type="checkbox"/> Other items or information:</p>			
Application Data Sheet, One sheet of Formal Drawings showing Figure 1 and return postcard.			

U.S. APPLICATION NO. (if known) <b>10/088186</b>		INTERNATIONAL APPLICATION NO. <b>PCT/KR00/01039</b>		ATTORNEY'S DOCKET NUMBER <b>5294-000010</b>	
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21. ☒ The following fees are submitted:

**BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):**  
 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... **\$1040.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... **\$890.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... **\$740.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(l)-(4) ..... **\$710.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(l)-(4) ..... **\$100.00**

**ENTER APPROPRIATE BASIC FEE AMOUNT =**

Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(e)).

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	13 - 20 =	0	x \$18.00	\$ 0.00
Independent claims	5 - 3 =	2	x \$84.00	\$ 168.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ \$280.00
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 1,488.00

☒ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2. + 744.00

**SUBTOTAL =** \$ 744.00

Processing fee of **\$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(f)). \$ 0.00

**TOTAL NATIONAL FEE =** \$ 744.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **\$40.00** per property + 0.00

**TOTAL FEES ENCLOSED =** \$ 744.00

	Amount to be refunded:	\$
	charged:	\$

a. ☒ A check in the amount of \$ 744.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 08-0750 A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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 REGISTRATION NUMBER  
 Dated: March 15, 2002

1/pst

**A NOVEL AROMATIC POLYSULFIDE AND AN ASPHALT  
COMPOSITION CONTAINING THE SAME**

**5 BACKGROUND OF THE INVENTION**

**Field of the Invention**

The present invention relates to a novel aromatic polysulfide and an asphalt composition containing the same. More particularly, this invention relates to a novel aromatic polysulfide and an asphalt composition, an adhesive  
10 promoter and an ultraviolet absorber containing the same.

**Description of the Related Art**

Korean Patent Application No. 1993-14434 discloses a modified asphalt composition comprising 0.5-100 wt% of polyolefin-type metal complex based  
15 on the 100 wt% of asphalt in order to improve physical properties of prior asphalt composition.

Korean Patent Application No. 1996-81058 reports a novel asphalt modifier with lower viscosity and odor-free character and an asphalt composition containing the above asphalt modifier, prepared in such a manner that  
20 naphthenic acid containing sulfur compound and other impurities is separated by distillation at the temperature of 70-200°C and then remaining volatile portions are removed by passing inert gas, after which oils such as lubricating oil are added to the treated compound. Korean Patent Application No. 1992-8279 also sets forth a cold-mixed asphalt paving composition containing  
25 straight asphalt, gas oil and polymeric hardening agent.

Meanwhile, U.S. Patent Nos. 4,244,747, 4,234,346 and 4,801,332 disclose an improved asphalt composition further comprising organic metal compound

such as organic manganese compound, organic cobalt compound. Furthermore, U.S. Patent No. 4,008,095 describes an asphalt paving composition comprising microgranular undigested coal particles, bituminous coal and asphalt.

5 An asphalt composition comprising polyolefins modified with a carboxyl group and/or other functional group derived from the carboxyl group is described in Japanese Patent Kokai Nos. Sho 54-139925, Sho 59-138263, Sho 60-158256, Sho 62-181358 and Sho 62-275160.

U.S. Patent No. 5,710,196 provides an asphalt composition comprising a  
10 graft copolymer, for example, acrylonitrile-butadiene-styrene copolymer resin so as to enhance the properties of asphalt composition. Furthermore, in order to reduce the cracking of asphalt, especially at low temperatures due to heavy loads, U.S. Patent Nos. 4,547,399, 4,835,199 and 5,002,987 suggest compositions of elastomeric copolymers and asphalt.

15 However, the conventional asphalt compositions including the aforementioned compositions fail to meet the requirement for excellent asphalt, for example, adhesiveness, water resistance, cracking-durability and abrasion resistance.

In the meantime, copolymer of metacrylic ester mono and vinyl monomer,  
20 which has the chromophore group representing different ultraviolet (hereinafter referred to as "UV") absorption patterns from the metacrylic ester mono is provided as an UV absorber in Korean Patent Application No. 1988-700951. In addition, Korean Patent Application No. 1995-3408 discloses a novel bezazol compound designed for the UV absorber, prepared in such a manner  
25 that carboxylic derivatives and o-phenyldiamine are reacted at the temperatures ranging between 20°C-90°C in the presence of chloro sulfonic acid.

In an effort to reduce the detrimental effect of UV by absorbing UV, Korean Patent Application No. 1989-18583 provides conjugated bis-1,3-diketone derivatives of benzene and Korean Patent Application No. 1989-19891 provides a composition comprising oligomer of cyclohexenilidene cyanoacetate.

5

### SUMMARY OF THE INVENTION

To solve the long-felt need in the art, the inventor has made intensive studies and as a result, developed a novel aromatic polysulfide polymer, noting that the aromatic polysulfide polymer may improve various properties of asphalt composition and serve as excellent an UV absorber.

10

Accordingly, an object of this invention is to provide a novel aromatic polysulfide polymer and a method for preparing the same.

Another object of this invention is to provide an asphalt composition comprising the above polysulfide.

15

Still another object of this invention is to provide an adhesion promoter applied to polymer resin and an UV absorber.

Other objects and advantages of this invention will become more apparent from the detailed description to follow taken in conjunction with the appended claims.

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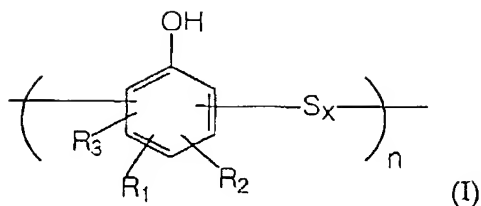
### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a graph representing the UV transmission pattern of the present polysulfide.

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### DETAILED DESCRIPTION OF THE INVENTION

This invention relates to a novel aromatic polysulfide having repeating units of the following formula (I):



5            wherein R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group; x is an integer of 1-4; and n is an integer of 2-10,000.

10            The structural feature of the present aromatic polysulfide is that elements in the 6B group, oxygen and sulfur, which have two electron lone pairs, are bonded to benzene nucleus. The electron lone pairs of oxygen and sulfur can simultaneously interact with cations, which may be components of asphalt paving composition, through electrostatic interaction, thereby improving a variety of physical properties of an asphalt paving composition.

15            The benzene ring of the polysulfide provides an ability to serve as excellent an UV absorber.

20            The average molecular weight of the present polysulfide may vary depending on polymerization, but preferably in the range of 5,000 to 20,000, and more preferably in the range of 7,000 to 15,000. If the molecular weight of the polysulfide is less than 5,000, an asphalt composition and an asphalt paving composition containing such polymer may be worse in view of durability, though inducing better processability. If the molecular weight is more than 20,000, the processability may be decreased, though temperature susceptibility and durability are increased.

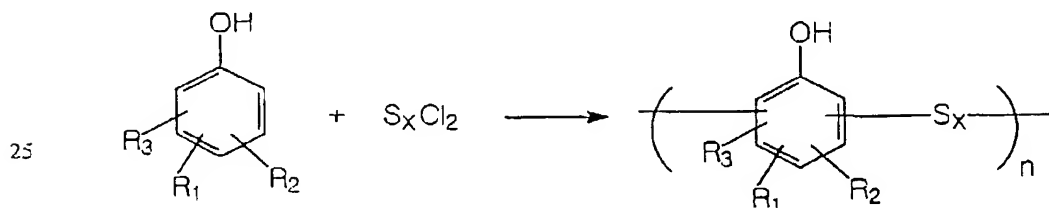
25            The substituents in the aromatic polysulfide of this invention may be any substituents, but preferably including unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group.

As used herein, "alkyl" means a branched or unbranched saturated hydrocarbon chain, which can be unsubstituted or substituted. For example, C<sub>1</sub>-C<sub>6</sub> straight or branched alkyl hydrocarbon chain contains 1 to 6 carbon atoms, and includes but is not limited to substituents such as methyl, ethyl, propyl, iso-propyl, butyl, iso-butyl, tert-butyl, n-pentyl, n-hexyl, and the like, unless otherwise indicated. The substituted alkyl has substituent(s) at one or more positions selected from halo, nitro, hydroxyl, alkyl, alkeyl, alkoxy, alkenyloxy, phenoxy, benzyloxy or aryl.

The term "aryl", alone or in combination, is defined herein as a monocyclic or polycyclic group, preferably a monocyclic or bicyclic group, i.e. phenyl or naphthyl, which can be unsubstituted or substituted, for example, with one or more and, in particular, one to three substituents selected from halogen, alkyl, hydroxy, alkoxy, haloalkyl, nitro, amino, acylamino, alkylthio, alkylsulfinyl and alkylsulfonyl. Some exemplary aryl groups include phenyl, 2-chlorophenyl, 3-chlorophenyl, 4-chlorophenyl, 2-methylphenyl, 4-methoxyphenyl, 3-trifluoromethylphenyl, 4-mitrophenyl, and the like.

This invention also relates to a method for preparing a novel aromatic polysulfide represented by the following scheme (I), comprising the step of reaction between phenol analogue and sulfur chloride species:

Scheme (I)



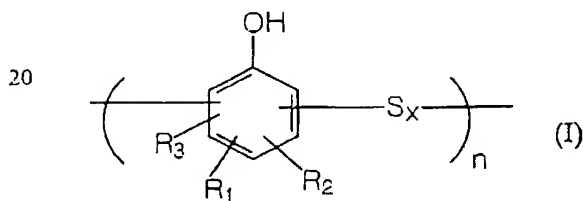
wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and

independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group; x is an integer of 1-4; and n is an integer of 2-10,000.

As explained above, the method of the present invention is performed by the reaction of phenol or phenol derivatives and sulfur chloride species, which may be classified as condensation polymerization, and the reaction condition is mild. For instance, the preferred reaction temperature is in the range of 50°C-90°C.

In addition, it is preferred that the method of this invention further comprises the step of adding an alkyl halide or aryl halide so as to add various substituents into the benzene nucleus. According to the method of this invention, the sulfur chloride species used is selected from the group consisting of sulfur dichloride, sulfur monochloride, trisulfur dichloride and tetrasulfur dichloride, and the most preferred sulfur chloride species is sulfur monochloride.

This invention provides an asphalt composition comprising: (i) a novel aromatic polysulfide having repeating units of the following formula (I):



wherein R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group; x is an integer of 1-4; and n is an integer of 2-10,000; and (ii) an asphalt.

The aromatic polysulfide contained in the asphalt composition is



responsible for improving various properties such as penetration, ductility, and temperature susceptibility. According to the preferred example, the amount of the polysulfide compound is from 0.5 to 10 wt% and the amount of the asphalt is from 90 to 99.5 wt% based on the weight of the composition. If  
 5 the amount of the polysulfide is less than 0.5 wt%, the effect of adding polysulfide may be negligible, but in case of exceeding 10 wt%, cracking is caused by hardening of the composition. Furthermore, if the amount of asphalt composition is less than 90 wt%, the fluidity may be decreased, but in excess of 99.5 wt%, the durability may be dramatically decreased though strength is  
 10 increased.

This invention also relates to an asphalt paving composition comprising an asphalt composition aforementioned (comprising 0.5 to 10 wt% of polysulfide having repeating units of formula (I) and 90 to 99.5 wt% of asphalt), aggregate,  
 15 stone powder and sand. According to the preferred example, the asphalt paving composition comprises 4 to 10 wt% of the asphalt composition, 65 to 85 wt% of the aggregate, 3 to 10 wt% of the stone powder and 8 to 25 wt% of the sand.

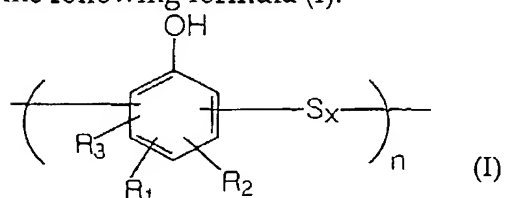
According to this invention, an useful asphalt can be any conventional  
 20 asphalt in the art, and preferably includes straight asphalt, blown asphalt, lake asphalt, rock asphalt, sand asphalt, asphaltite, etc., and the most preferably includes straight asphalt.

In the asphalt paving composition of this invention, if the amount of the asphalt composition is less than 4 wt%, the physical strength and the fluidity  
 25 may be remarkably decreased, but in case of exceeding 10 wt%, the durability may be decreased.

The asphalt paving composition exhibits excellent adhesiveness to

aggregate component and better water resistance, and renders the temperature susceptibility less sensitive, thereby greatly improving durability.

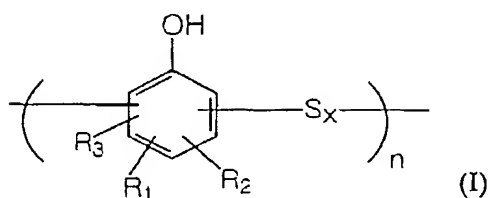
This invention also provides an adhesion promoter applied to polymer resin, which comprises a novel aromatic polysulfide having repeating units of the following formula (I):



wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group;  $x$  is an integer of 1-4; and  $n$  is an integer of 2-10,000.

The aromatic polysulfide serving as an adhesion promoter renders polymer resin (for example, polyethylene, polypropylene, polyisobutylene, polyvinylchloride, polystyrene, polyvinylacetate, polyisoprene) containing the polysulfide to be more adhesive.

This invention provides an UV absorber comprising a novel aromatic polysulfide having repeating units of the following formula (I):



wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group,

unsubstituted aryl group or substituted aryl group; x is an integer of 1-4; and n is an integer of 2-10,000.

The aromatic polysulfide may function as an UV absorber in paint, plastic film, etc. to reduce sensitivity to UV, thereby extending the life span of such product.

The following specific examples are intended to be illustrative of the invention and should not be construed as limiting the scope of the invention as defined by the appended claims.

#### EXAMPLE 1 : Preparation of the Aromatic Polysulfide of this Invention I

In a 5-necked flask of 5 liters fitted with 2-additional funnels, water cooled condenser, agitator and thermometer was placed *para*-cresol (2160 g: 20 mole) and heated to 70°C . To it was added sulfur monochloride (2835 g: 21mole) dropwise while maintaining the reaction temperature at 50°C-70°C . When 70% of the sulfur monochloride was added, methylene chloride (1000 ml) was introduced. Then, the remaining sulfur monochloride was added, and heated for an additional hour at 70°C-80°C .

Upon completion of the reaction, 2000 ml of water was added to remove any unreacted sulfur monochloride. Removal of solvent and water gave a dark yellowish solid polymer.

The resultant polymer shows about 8,000 of the average molecular weight and 80°C of the melting point. The IR absorption spectrum of the polymer exhibits troughs at 3400 cm<sup>-1</sup> , 1440 cm<sup>-1</sup> , 1220 cm<sup>-1</sup> and 1160 cm<sup>-1</sup>.

#### EXAMPLE 2 : Preparation of the Aromatic Polysulfide of this Invention II

The aromatic polysulfide according to this invention was prepared in the

same manner as in EXAMPLE 1, except that phenol was used instead of *para*-cresol.

The final product shows about 8,000 of the average molecular weight and 80 °C of the melting point.

5

**EXAMPLE 3 : Preparation of the Aromatic Polysulfide of this Invention III**

The aromatic polysulfide according to this invention was prepared in the same manner as in EXAMPLE 1, except that *meta*-cresol was used instead of *para*-cresol.

10 The yielded product shows about 8,000 of the average molecular weight and 80 °C of the melting point.

**EXAMPLE 4 : Preparation of the Aromatic Polysulfide of this Invention IV**

15 The aromatic polysulfide according to this invention was prepared in the same manner as in EXAMPLE 1, except that mixed cresol (including *ortho*-cresol, *para*-cresol and *meta*-cresol) was used instead of *para*-cresol.

The yielded product shows about 8,000 of the average molecular weight and 80 °C of the melting point.

20 **EXAMPLE 5 : Preparation of the Asphalt Composition of this Invention**

1 wt%, 2 wt% and 3 wt% of aromatic polysulfide prepared in EXAMPLE 1 were respectively added to 99 wt%, 98 wt% and 97 wt% of straight asphalt, thereby making total weight percent of the final composition 100 wt%. Then while melting, each mixture was completely mixed to yield the asphalt  
25 composition at 150 °C .

**EXAMPLE 6 : Preparation of the Asphalt Paving Composition of this Invention**

6 wt% of each asphalt composition prepared in EXAMPLE 5, 34 wt% of aggregate having 19-13 mm of particle size, 40 wt% of aggregate having less than 13 mm of particle size, 15 wt% of sand and 5 wt% of stone powder were heated and mixed to yield the asphalt paving composition at 150°C .

**EXPERIMENTAL EXAMPLE 1 : Evaluation on Properties of the Asphalt Composition of this Invention**

The various physical properties (including penetration, ductility, softening temperature and index of temperature susceptibility) of the asphalt composition prepared in EXAMPLE 5 were evaluated based on KS M 2252, KS M 2254 and KS M 2250, respectively. The results are summarized below Table 1:

**EXPERIMENTAL EXAMPLE 2 : Evaluation on Properties of the Asphalt Paving Composition of this Invention**

The Marshall test on asphalt paving composition prepared in EXAMPLE 6 was carried out based on KS F 2337. And the Wheel tracking test on the asphalt paving composition prepared in EXAMPLE 6 was performed so as to evaluate resistance to rutting caused by wheels based on the Manual for Testing Qualities of Construction issued by the Korea Highway Corporation. The results are summarized below in Table 1:

TABLE 1

Items	Physical Properties		Amount of aromatic polysulfide(wt%)		
			1	2	3
Asphalt Composi- tion	Penetration (1/10mm, 25℃)		85	82	79
	Ductility (cm, 25℃)		150	150	150
	Index of temperature susceptibility (℃)		0.0263	0.0178	0.0161
	Softening temp.(℃)		59	67	72
Asphalt Paving Composi- tion	Marshall test	Stability(kg)	1.687	1.892	1.956
		Fluidity(1/100cm)	30	28	27
		Porosity(%)	4.02	4.51	4.78
	Wheel tracking	Ratio of deformation(mm/min)	0.035	0.014	0.009
		Dynamic stability(times/mm)	921	1237	1516

### EXPERIMENTAL EXAMPLE 3 : Evaluation on the Enhancing Adhesion of the Aromatic Polysulfide of this Invention

In order to evaluate the adhesion enhancement of the polysulfide, 3 wt% of the polysulfide prepared in EXAMPLE 1 were added to the PVC solution in THF (tetrahydrofuran), the resultant was then coated on a stainless steel plate and finally dried. Following the lapse of proper time, the extent of adhesion was observed with the naked eyes. It was noted that the PVC solution containing the polysulfide of this invention adhered strongly to the stainless steel plate, thus not observing peeling off, but the PVC solution without the polysulfide was peeled off from the plate with a lapse of time.

As a result, it is confirmed that the polysulfide of this invention may remarkably enhance the adhesion of polymer resin.

**EXPERIMENTAL EXAMPLE 4 : Evaluation on the UV Transmittance of the  
Aromatic Polysulfide of this Invention**

In an effort to evaluate the UV transmittance of the polysulfide prepared in EXAMPLE 1, 0.016 g of the polysulfide was dissolved in 8 ml of chloroform solution and then the UV transmittance was measured with UV-spectrophotometer (HP-8454). The transmittance was calculated in the following equation and the result was plotted as shown in Fig. 1:

**Equation (I)**

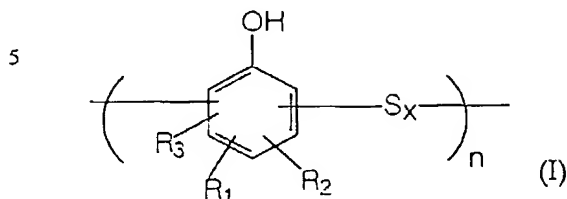
$$\text{Transmittance (T)} = 10^{-A(\lambda)}$$

wherein  $A(\lambda)$  is absorbance.

As shown in Fig. 1, the polysulfide of this invention represents a much lower transmittance in the UV region and therefore has excellent property as an UV absorber.

What is claimed is:

1. A novel aromatic polysulfide having repeating units of the following formula (I):

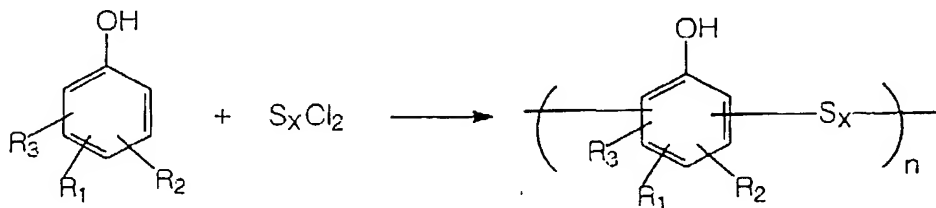


wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and  
 10 independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group;  $x$  is an integer of 1-4; and  $n$  is an integer of 2-10,000.

2. The novel aromatic polysulfide according to claim 1, wherein the average  
 15 molecular weight of the aromatic polysulfide is from 5,000 to 20,000.

3. A method for preparing a novel aromatic polysulfide represented by the following scheme (I), comprising the step of reaction between phenol or derivatives thereof and sulfur chloride species:

20 Scheme (I)



25

wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group,



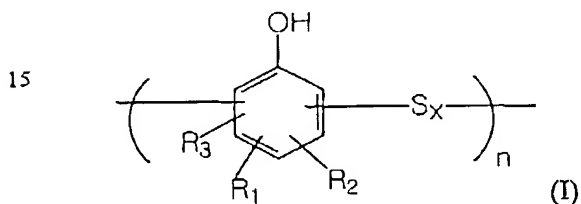


selected from the group consisting of straight asphalt, blown asphalt, lake asphalt, rock asphalt, sand asphalt and asphaltite.

9. An asphalt paving composition comprising the asphalt composition of claim 6, aggregate, stone powder and sand.

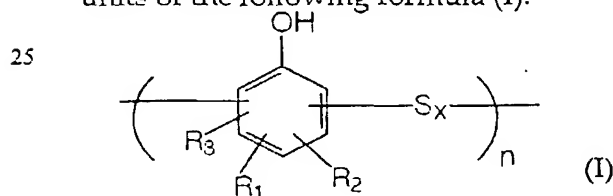
10. The asphalt paving composition according to claim 9, wherein the amount of the asphalt composition is from 4 to 10 wt%, the amount of the aggregate is from 65 to 85 wt%, the amount of the stone powder is from 3 to 10 wt% and the amount of the sand is from 8 to 25 wt%.

11. An adhesion promoter applied to polymer resin, which comprises a novel aromatic polysulfide having repeating units of the following formula (I):



wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group;  $x$  is an integer of 1-4; and  $n$  is an integer of 2-10,000.

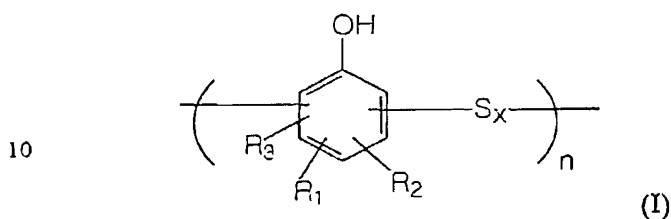
12. An UV absorber comprising a novel aromatic polysulfide having repeating units of the following formula (I):



wherein  $R_1$ ,  $R_2$  and  $R_3$  are the same or different from each other, and independently represent H, unsubstituted alkyl group, substituted alkyl group, unsubstituted aryl group or substituted aryl group;  $x$  is an integer of 1-4; and  $n$  is an integer of 2-10,000.

# ABSTRACT

The present invention relates to a novel aromatic polysulfide and an asphalt composition containing the same, and more particularly, to a novel aromatic polysulfide having repeating units of the following formula (I), an asphalt composition, an asphalt paving composition, an adhesion promoter and an UV absorber containing the same:



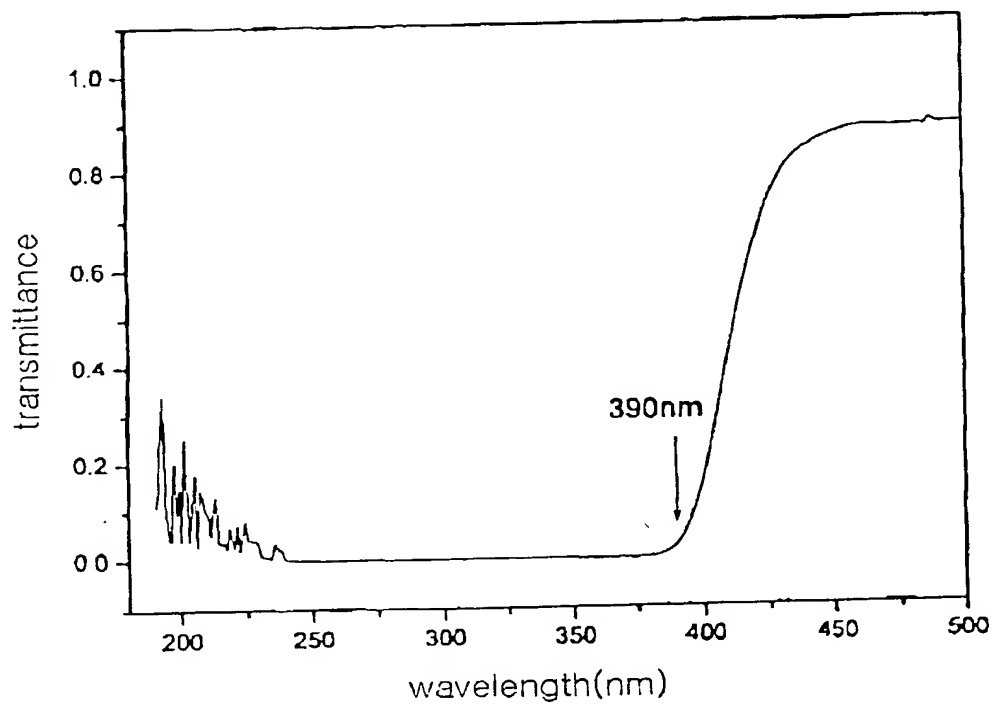
The asphalt paving composition of this invention exhibits excellent adhesiveness to aggregate component and better water resistance, and renders the temperature susceptibility less sensitive, thereby greatly improving durability.

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## FIGURE

Fig. 1



# Declaration and Power of Attorney for Patent Application

## 특허 출원 관련 선언 및 위임권

### Korean Language Declaration

아래 지명된 발명자로서, 본인은 하기 사항을 선언합니다.

본인의 거주지, 우송 주소 및 국적은 본인의 성명 아래에 기재된 것과 동일합니다.

본인은 하기 명시된 발명에 대한 독점권 청구하는 주체의 최초 원예 단독 발명자이거나 (아래에 한 이름만이 기재된 경우) 또는 최초 원예 공동 발명자임을 (아래에 여러 이름이 기재된 경우) 확인합니다.

A NOVEL AROMATIC POLYSULFIDE AND AN  
ASPHALT COMPOSITION CONTAINING THE SAME

다음 난이 체크되어 있지 않으면 본 발명의 명세서가 여기에 첨부됩니다.

☒ 미합중국 출원번호 또는 PCT 국제 출원번호는  
September 15, 2000 로  
PCT/KR00/01039 입예 출원되었고  
입예 개정되었음  
(해당 경우).

본인은 상기 개정에 의해 수정된 상기 명세서는 물론 특허 청구의 내용을 검사했으며 이해했음을 확인합니다.

본인은 연방 규정 코드인 제37장의 제1.56항에 의거하여 특허 자각에 관한 자료 정보를 공개할 의무를 인정합니다.

As a below named Inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

A NOVEL AROMATIC POLYSULFIDE AND AN  
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the specification of which is attached hereto unless the following box is checked:

☒ was filed on September 15, 2000  
as United States Application Number or PCT  
International Application Number  
PCT/KR00/01039 and was amended on  
                     (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

### Korean Language Declaration

본인은 외국인 특허 출원(들)이나 발명자의 증명서 관련 경우에는 미합중국 코드인 제35장의 제17.9(a)-(d)항이나 제365(b)항에 의거하여 또는 미합중국 이외에 적어도 한 국가를 지칭하는 PCT 국제 출원의 경우에는 제365(a)항에 의거하여 하기 명시된 특허 출원의 외국 우선권을 주장하며, 외국인 특허 출원, 발명자 증명서 또는 우선권이 주장되는 출원일 이전에 제출된 PCT 국제 출원도 또한 여기에 해당함을 체크함으로써 확인하였습니다.

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed

Priority Not Claimed  
우선권 주장 없음

1999-39522  
(Number)  
(번호)

Korea  
(Country)  
(국가)

September 15, 1999  
(Day/Month/Year Filed)  
(출원일자 월/일/년)

☐

(Number)  
(번호)

(Country)  
(국가)

(Day/Month/Year Filed)  
(출원일자 월/일/년)

☐

본인은 미합중국 코드인 제35장 제119항(c)에 명시된 바와 같이 하기 미합중국 가출원에 관련된 특권을 요구합니다.

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below

(Application No.)  
(출원 번호)

(Filing Date)  
(출원일자)

(Application No.)  
(출원 번호)

(Filing Date)  
(출원일자)

본인은 미합중국 코드인 제35장의 미국인 출원(들) 관련 제120항에 명시된 바와 같이 또는 미합중국을 지칭하는 PCT 국제 출원 관련 제365(c)항에 명시된 바와 같이 하기 출원의 특권을 요구합니다. 이 출원서에 있는 각 특허 청구의 내용은 미합중국 코드인 제35장 제112항의 첫번째 절에서 명시된 바와 같이 종전외 미국 또는 PCT 국제 출원에 발표되지 않았으면 본인은 연방 규정 코드인 제37장 제1.56항에 명시된 바와 같이 종전 출원일자와 이 출원서의 국내 또는 PCT 국제 출원일자 사이에 특허 자격에 대한 자료 정보를 공개할 의무를 인정합니다.

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application

(Application No.)  
(출원 번호)

(Filing Date)  
(출원일자)

(Status) (patented, pending, abandoned)  
(현황)(특허 획득, 출원중, 포기)

(Application No.)  
(출원 번호)

(Filing Date)  
(출원일자)

(Status) (patented, pending, abandoned)  
(현황)(특허 획득, 출원중, 포기)

본인이 아는 한도 내에서 여기에 제공된 모든 내용이 사실이고, 제공된 정보나 소신이 모두 사실임을 확인하며, 더 나아가 미합중국 코드 제18장의 제1001절에 명시된 바와 같이 그외의 허위 진술 및 이와 유사한 형식적 발급이나 부속으로 처벌 받거나 벌금과 감옥형을 모두 받을 수 있고 이러한 그외의 허위 진술은 특허 출원이나 후에 발급된 특허의 유효성을 위태롭게 함을 인지하면서 여기에 진술함을 선언합니다.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application of any patent issued thereon

### Korean Language Declaration

위임권: 지명된 발명자로서 본인은 이 특허를 증원하고 이와 관련하여  
특허 및 상표청이 요구하는 사무를 처리하기 위해서 하기 변호사(들)  
및/또는 대리인(들)을 임명합니다. (성명 및 등록번호 기입)

POWER OF ATTORNEY: As a named inventor, I hereby  
appoint the following attorney(s) and/or agent(s) to prosecute  
this application and transact all business in the Patent and  
Trademark Office connected therewith: (list name and  
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만약 있으면 두번째 공동 발명자의 이름	Full name of second joint inventor, if any
두번째 발명자의 서명 일자	Second Inventor's signature Date
거주지	Residence
국적	Citizenship
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그들의 서명을 제공함 것.)

(Supply information and signature for third and subsequent joint  
inventors)